Strain-controlled correlation effects in self-assembled quantum dot stacks

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We show that elastic interactions of an array of self-assembled quantum dots in a parent material matrix are markedly distinct from the elastic field created by a single point defect, and can explain the observed abrupt correlation–anticorrelation transition in semiconductor quantum dot stacks. Finite volume effects of the quantum dots are shown to lead to sharper transitions. Our analysis also predicts the inclination angle under which the alignment in successive quantum dot layers occurs in dependence on the material anisotropy.

The self-assembled growth of semiconductor quantum dots (QDs) is widely used as an efficient tool for fabricating nanostructures with important potential applications in opto- and nanoelectronics [1, 2]. The process of self-assembly is based on the formation of coherent localized structures confined in three spatial directions, i.e. QDs, on a flat, uniform two-dimensional (2D) wetting layer in highly strained materials, using the Stranski-Krastanov growth mode. Multilayer systems in the form of vertical stacks are used to improve the lateral ordering of QD arrays, and minimize the size and shape fluctuations of the QDs. By tuning the spacer thickness a certain desired lateral spacing of QDs can be obtained, and one can switch between several types of stacking modes [3, 4, 5, 6]. Theoretically, several approaches have been made to understand the new features of QD stacks. Elasticity theory has proven to be very useful, not only for understanding the growth of QD systems consisting of one layer [7], in particular when combined with kinetic Monte Carlo simulations [8, 9, 10], but also for giving insight into the growth of stacked QD systems [3, 4, 11, 12, 13, 14]. Our understanding of the vertical and lateral correlation and anticorrelation properties of such stacked arrays of QDs is, however, still incomplete. Recently, clear experimental evidence of an abrupt transition between vertically aligned (correlated) and antialigned (anticorrelated) QDs in the InGaAs/GaAs material system in dependence upon the spacer layer thickness has been presented [6], but this transition could not be explained by the anisotropic strain field of a single QD [4, 5]; and the angle against the vertical under which the alignment in successive QD layers is observed seriously disagrees with the theoretically predicted tilt angle for the minima of the strain [5].

In this report we present a three-dimensional model of the strain field of a stacked array of quantum dots, taking fully into account the anisotropy of the material system and the 3D shape of the QDs. This is an extension of the model proposed by Shchukin *et al.* [12]. While other theoretical descriptions [4, 12] using a similar Green's tensor approach [15] have employed strictly periodic arrays and point-like or stripe-like approximations of the QDs, we have implemented arbitrary QD shapes and configurations. Within our approach we are able to explain both the experimentally investigated tilt angles and positions in an anticorrelated stacked array of QDs in the InGaAs/GaAs material system [6] and describe the transition from correlated to anticorrelated growth.

We model the multi-sheet array of QDs by inclusions of material A in a matrix of material B. Starting from elasticity theory for anisotropic materials [12, 16], we solve the equilibrium equation $\nabla_j \left(\lambda_{ijkl} \nabla_k u_l(\mathbf{r}) \right) = \nabla_j (\sigma_{ij}^{(0)} \vartheta(\mathbf{r}))$ with the elastic moduli λ_{ijkl}, the elastic displacement field u_l, the characteristic shape function $\vartheta(\mathbf{r})$ of the QDs ($\vartheta(\mathbf{r}) = 1$ if $\mathbf{r}$ is inside an inclusion and zero elsewhere), and the stress tensor $\sigma_{ij}^{(0)}$ which is connected to the stress-free strain tensor $\varepsilon_{ij}^{(0)}$ by $\sigma_{ij}^{(0)} = \lambda_{ijkl} \varepsilon_{kl}^{(0)}$ [16]. Here Einstein's summation convention is used. Since the volume of the inclusions is assumed to be small compared to the parent material, the homogeneous moduli approximation is used. The problem is solved under stress-free boundary conditions on the surface.

In general, the elastic energy of interacting inclusions is given by [16]

$$E_{el} = \frac{1}{2} \int d^3\mathbf{r} \sigma_{ij}^{(0)} \vartheta(\mathbf{r}) \left(\varepsilon_{ij}^{(0)} - \varepsilon_{ij}(\mathbf{r}) \right). \qquad (1)$$

Introducing the in-plane coordinates $\mathbf{r}_\parallel$ and the vertical coordinate z, one can express the elastic interaction energy per unit surface area in the growth plane at $z = z_0$ in terms of the static Green's tensor as [12]

$$E(\mathbf{r}_\parallel; z = z_0) = h^{(S)} \int d^2\mathbf{r}'_\parallel \int_{-\infty}^{z_0} dz' \vartheta(\mathbf{r}'_\parallel; z') W(\mathbf{r}_\parallel - \mathbf{r}'_\parallel; z_0 - z') \qquad (2)$$

where the integral is over the buried QDs with $z' \leq z_0$, $h^{(S)}$ is the height of one single inclusion on the surface, and the interaction term

$$\begin{aligned} W(\mathbf{r}_\parallel - \mathbf{r}'_\parallel; z_0 - z') &= \int \frac{d^2\mathbf{k}_\parallel}{(2\pi)^2} e^{i\mathbf{k}_\parallel (\mathbf{r}_\parallel - \mathbf{r}'_\parallel)} \\ &\times \left\{ \sigma_{ij}^{(0)} \left[\nabla_j \nabla'_m \tilde{G}_{il}(\mathbf{k}_\parallel; z_0, z_0 - z') \right] \sigma_{lm}^{(0)} \right\} \end{aligned} \qquad (3)$$

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is related to the Fourier transform of the static Green's tensor $\tilde{G}_{il}$, which can be computed numerically for elastically anisotropic cubic crystals, for details see [12, 15, 16, 17].
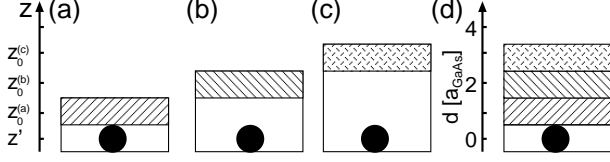

FIG. 1: Schematic representation of the dependence of the surface elastic energy upon the spacer thickness. The cross-sections of samples with different spacer thickness d (a)-(c) are combined into a single cross-section (d).
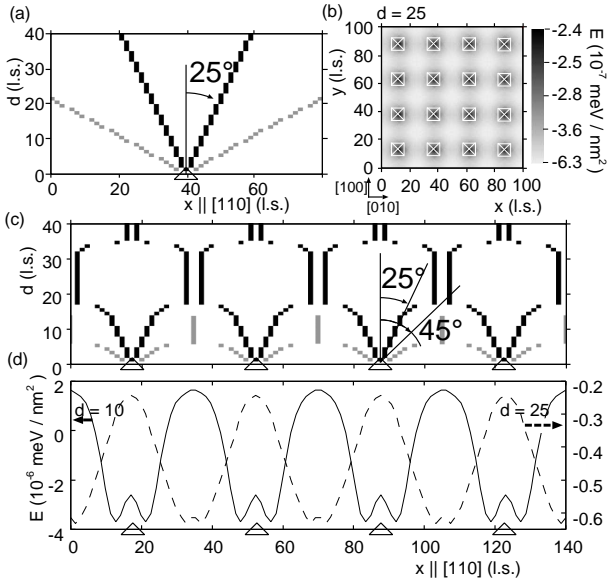

FIG. 2: Elastic strain field of one single point-like inclusion (a) and a periodic array of point defects (b)-(d) in a GaAs matrix. (a) and (c) show the minima (black) and maxima (grey) of the elastic energy E per unit area in the [110] direction of the relaxed surface vs. spacer thickness d. The density plot (b) depicts E in the [001]-[010] surface plane at $d = 25$. (d) E for $d = 10$ and $d = 25$. The positions of the QDs are indicated by triangles (a),(c),(d) and by boxes (b), respectively. All lengths are in units of the lattice constant of the parent material $a_{GaAs} = 0.565$ nm (monolayer width).

We have computed the elastic energy in the relaxed surface at $z = z_0$ for a given array of buried QDs at $z = z'$ in dependence on the spacer thickness $d = z_0 - z'$. The results are combined into a single cross-sectional plot as schematically illustrated in Fig. 1. Periodic boundary conditions in the lateral direction are used. Fig. 2(a) shows the minima (black) and the maxima (grey) of the elastic interaction energy E in the relaxed surface for a single buried point-like QD. The minima and maxima occur in the [110] direction at angle $\alpha = 25°$ and $\tilde{\alpha} = 60°$ against the vertical, and correspond to maximum tensile
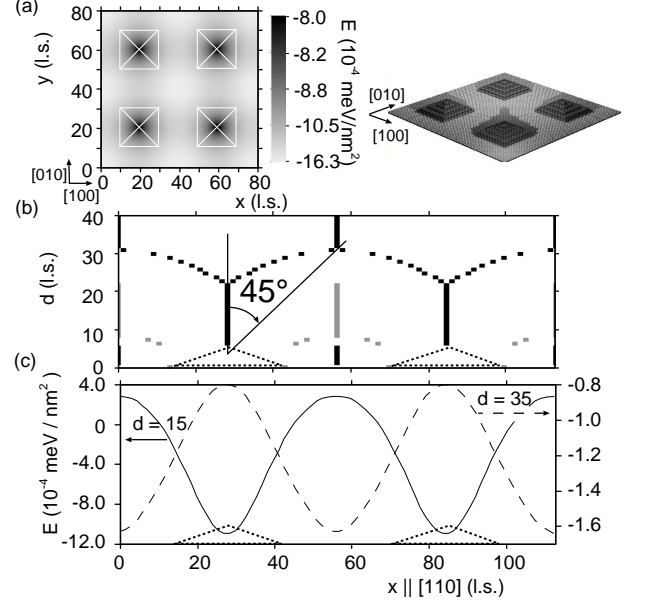

FIG. 3: Elastic strain field of a periodic array of pyramids in a GaAs matrix. (a) density plot of the elastic energy per unit area E in the [001]-[010] surface plane at $d = 35$ (inset: 3D view) (b) minima (black) and maxima (grey) of E in the [110] direction of the relaxed surface vs. spacer thickness d. (c) E for $d = 15$ and $d = 35$. The positions and sizes of the QDs are indicated by boxes (a) and dotted triangles (b),(c).

strain ($E < 0$, black) and maximum compressive strain ($E > 0$, grey), respectively. Regions of tensile strain give rise to an attractive potential and act as preferred nucleation centers for QDs, whereas regions of compressive strain form repulsive potentials [18].

As can be seen, our model is in good agreement with previous theoretically predicted values of the angle α in which the minima of the elastic energy in the [110]-[001]-plane occur [4]. Computing the inclination angle α for other material systems, we find that α increases with increasing anisotropy ratio $A = 2c_{44}/(c_{11} - c_{12})$ of the parent material, where c_{11}, c_{12}, c_{44} are the elastic moduli in Voigt notation. We obtain $\alpha = 19°$ for Si ($A = 1.56$), $\alpha = 25°$ for GaAs ($A = 1.83$), and $\alpha = 33°$ for ZnSe ($A = 2.04$), in agreement with [4]. However, these results contradict the experimental observation of much larger angles [6].

The situation changes drastically if we consider an array of QDs (Fig. 2(b)-(d)). We use a quadratic array of 4×4 QDs with a lateral spacing of $l = 25$ lattice sites (l.s.). In a neighborhood of approximately $l/2$ around each QD the elastic properties are governed only by this single point defect. Outside this region the strain fields are overlapping, which leads to new effects (Fig. 2(c)). Starting with a spacer thickness of $d \approx 14$ l.s., the inclination angle α increases more and more until at $d = 17$ l.s. the two minima induced by two neighboring QDs meet and form a flat double minimum exactly in the middle between these QDs. With further increasing spacer thick-

ness the position of the double minimum remains stable and unchanged for a considerable range of d-values. Finally, at $d = 31$ l.s the minimum starts moving back to a position vertically above the QDs.

The transition of the minima from a position close to vertically above the QDs to in between the QDs is also visible in the elastic energy profiles at $d = 10$ l.s. and $d = 25$ l.s. in Fig. 2(d). This indicates a transition from correlated to anticorrelated growth. From Fig. 2(c) we infer an inclination angle for alignment of QDs of about $\alpha = 45°$ for GaAs which is in reasonable agreement with the experimentally observed value of $50°$ [6]. The small remaining discrepancy might result from several subtle effects. First, in our model the material of the QDs is completely neglected. Second, stacking of the QDs might also be influenced by modulations in chemical composition or be due to morphological changes in the spacer layer.

For different materials, again the inclination angle α increases with increasing anisotropy, as in the case of one single QD: $\alpha = 43°$ for Si, $\alpha = 45°$ for GaAs, and $\alpha = 47°$ for ZnSe.

We have also studied the influence of the shape and volume of the QDs. In Fig. 3 we consider a 2×2 array of pyramids, each with a baselength of 20×20 l.s. and a height of 5 l.s positioned with a lateral distance of $l = 40$ l.s. Compared with an array of defects, there are significant differences. The inclination angle α remains unchanged. But the finite volume of the QDs results in a large range of spacer thickness d where the minimum energy is directly located vertically above the buried structures (Fig. 3 (b)). At a spacer thickness of about four times the height of the pyramids the energy minimum splits and moves towards the anticorrelated positions between the QDs. Due to this finite size effect, the transition from the correlated to the anticorrelated regime happens much more abruptly than in an array of point

defects (Fig. 2(c)). Such an abrupt transition is indeed observed experimentally [6]. Also the double minimum of the energy profile is replaced by a single minimum (Fig. 3(c)) . Note that the transition occurs at larger spacer thickness $d \approx 32$ for the array of QDs with a larger lateral distance of $l = 40$ l.s., and this ratio should scale for arrays with even larger lateral distance at fixed angle α. This is consistent with the experiment [6] where the transition was observed at a spacer thickness of 150 monolayers for an in-plane nearest-neighbor distance of 80–100 nm.

In conclusion, we have shown that elastic interactions of an array of QDs in a parent material matrix explain the observed correlation–anticorrelation transition, in contrast to calculations which take into account only a single point defect. The observed material trend of increasing inclination angles of the vertical stacking mode for larger anisotropy is also corroborated. The abrupt transition has been attributed to the finite volume of the QDs, and the reclining angle under which the anticorrelated QDs align is in good quantitative agreement with the experiment on self-organized InGaAs/GaAs quantum dot stacks.

The algorithm introduced in this paper allows for fast computation of the elastic interaction energy of nanostructures with arbitrary shape and spatial arrangement in three dimensions, and the only parameters are the elastic moduli of the considered anisotropic cubic parent material. Thus it is appropriate for implementation into kinetic Monte Carlo simulations [10, 13] which might be used for a detailed investigation of the growth kinetics of self-assembled QD stacks in future research.

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